Electronic Structure Calculations with Dynamical Mean Field Theory in δ-Pu

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Plutonium (Pu) exhibits complex and anomalous properties, including several structure phase transitions and large volume expansion. These unusual properties are largely due to the peculiar nature of electronic structure — its 5f electrons are delicately balanced between being itinerant and localized. Understanding these fundamental properties is a challenge in condensed matter physics. Traditional first-principle electronic structure calculations (based on the density functional theory) has found tremendous success in exploring the itinerant behaviors in simple metals and semiconductors while the dynamical mean field theory (DMFT) gives a nonperturbative treatment of the interplay between itinerant and localized behaviors in strongly correlated electronic systems. Recently there is increased interest in combining the first-principle approach with DMFT [1].

In view of the importance of this material to the Laboratory's mission, it is relevant for us to have such a computational capability. The success of this kind of approach relies on accurate and efficient density functional approaches and a powerful impurity solver. At the current stage, we are studying the electronic structure in δ-Pu by combining the DMFT with the recently developed tight-binding fitting method. The correlation effect is described by introducing an on-site Coulomb repulsion U on 5f orbitals as a model parameter. The solution comes with finding the self-consistency between the mapped impurity Green's function with the site-diagonal Green's function where the lattice effects enter. In the presence of U, the chemical potential is adjusted to have total number of 7s6p6d5f electrons to be 14. The calculation is carried out for the theoretical equilibrium volume of fcc structure by using the Hubbard-I approximation as an impurity solver.

In Fig. 1, we compare the total density of states for the case of U = 0 between the Green's function approach (black line) with an intrinsic lifetime broadening and direct diagonalization (green line) with a simple Fermi-Dirac temperature smearing. The energy is measured with respect to the Fermi energy. As is shown, the agreement is excellent. It means that in the absence of strong correlation, the Green's function approach recovers the results obtained by directly diagonalizing the Hamiltonian within the density functional theory.

In Fig. 2, we show the total density of states for the case of U = 4 eV from the Green's function approach. Note that both the lower and upper Hubbard bands are exhibited, as a typical feature of the strong correlation effect. It also shows that the Fermi energy cuts the edge of the lower Hubbard band. However, the Hubbard-I approximation, which is more suitable to describe the paramagnetic Mott insulator, cannot give the coherent peak at the Fermi energy as observed experimentally.

Therefore, other more sophisticated impurity solvers are needed, which we are working on now. Also, we are implementing the DMFT directly into the *ab-initio* electronic code, typically FP-LMTO, which is required for full first-principle calculations.

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[1] G. Kotliar et al., *Rev. Mod. Phys.*, **78** (April 1st, 2006).

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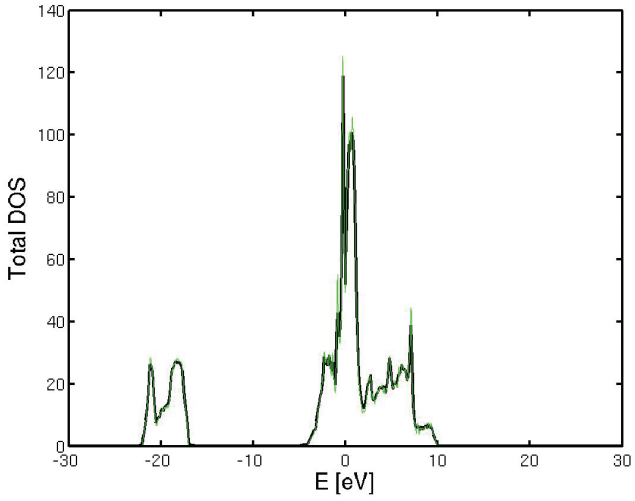

Fig. 1.
Total density of states for the case of U = 0 obtained with the Green's function approach (black line) and exact diagonalization (green line). Note that the results from two methods are in good agreement.

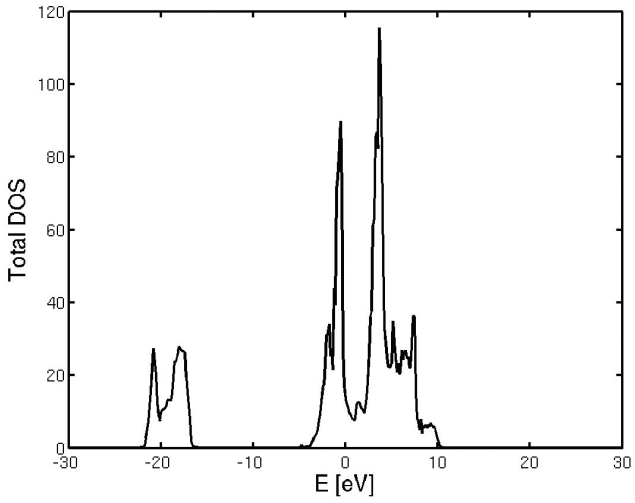

Fig. 2.
Total density of states for the case of U = 4 eV calculated with the Green's function approach. Typical of the strong correlation effect, the lower and upper Hubbard bands are exhibited.